

In an electric double layer capacitor, the presence of water in the organic electrolyte, the solvent or the pores of an electrode material, such as carbon, is detrimental for the performance and leads to decreased capacitance and increased internal resistance. If the moisture in the cells is electrolyzed to form a gas, the gas remains in the pores and expels electrolyte from the pores which will lower the capacitance, particularly after long-term use. It is therefore an object of the present invention to maintain the high capacity of an electric capacitor by preventing degrading of its performance due to presence of water. Accordingly, the present invention as set forth in **Claims 5, 12 and 20** relates to methods for producing electric double layer capacitors in which the organic electrolyte contains benzene or its chlorine derivative (specification, page 1-4). The benzene or its chlorine derivative have a high affinity to the pseudo-graphite surface present on the inner walls of pores of the carbonaceous material and are likely to be adsorbed instead of water in the fine pores. When a voltage is applied, the water can be electrolyzed and the gas will be present outside the fine pores and can be discharged as bubbles out of the element by reducing the pressure. In contrast, in conventional elements, having no benzene or chlorine derivative in the electrolyte, the gas generated by the electrolysis of water in the fine pores will remain in the fine pores, thereby deteriorating the performance of the capacitor (specification, page 6, line 7 to page 7, line 12).

Accordingly, the first embodiment of the present invention as claimed in **Claim 5** relates to a **method for producing an electric double layer capacitor**, which comprises impregnating an element comprising positive and negative electrodes facing each other with a separator interposed between them, with an organic electrolyte capable of forming an electric double layer on the surface of the electrodes to store electric charge, and then applying a voltage to the element, wherein said positive and negative electrodes are made of electrodes

containing a carbonaceous material having a specific surface area of from 100 to 3,000 m²/g, and said organic electrolyte contains benzene or its chlorine derivative having at least one hydrogen atom of benzene substituted by a chlorine atom;

wherein after the application of a voltage to the element, the element is maintained under reduced pressure;

wherein the organic electrolyte contains

a) a solvent selected from the group consisting of ethylene carbonate, propylene carbonate, butylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, acetonitrile, glutaronitrile and a mixture thereof; or

b) a solvent mixture of sulfolane and a solvent selected from the group consisting of ethylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, acetonitrile, glutaronitrile and a mixture thereof; or

c) a solvent mixture of a sulfolan derivative and a solvent selected from the group consisting of ethylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, acetonitrile, glutaronitrile and a mixture thereof.

Independent **Claim 12** relates to a **method for producing an electric double layer capacitor**, which comprises impregnating an element comprising positive and negative electrodes facing each other with a separator interposed between them, with an organic electrolyte capable of forming an electric double layer on the surface of the electrodes to store electric charge, and then applying a voltage to the element, wherein said positive and negative electrodes are made of electrodes containing a carbonaceous material having a specific surface area of from 100 to 3,000 m²/g, and said organic electrolyte contains benzene or its chlorine derivative having at least one hydrogen atom of benzene substituted by a chlorine

atom;

wherein the voltage is applied to the element in a dry atmosphere in an open condition, and thereafter, the element is maintained under reduced pressure;

wherein the organic electrolyte contains

a) a solvent selected from the group consisting of ethylene carbonate, propylene carbonate, butylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, acetonitrile, glutaronitrile and a mixture thereof; or

b) a solvent mixture of sulfolane and a solvent selected from the group consisting of ethylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, acetonitrile, glutaronitrile and a mixture thereof; or

c) a solvent mixture of a sulfolan derivative and a solvent selected from the group consisting of ethylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, acetonitrile, glutaronitrile and a mixture thereof.

Independent **Claim 20** relates to a **method for producing an electric double layer capacitor**, which comprises impregnating an element comprising positive and negative electrodes facing each other with a separator interposed between them, with an organic electrolyte capable of forming an electric double layer on the surface of the electrodes to store electric charge, and then applying a voltage to the element, wherein said positive and negative electrodes are made of electrodes containing a carbonaceous material having a specific surface area of from 100 to 3,000 m²/g, and said organic electrolyte contains benzene or its chlorine derivative having at least one hydrogen atom of benzene substituted by a chlorine atom;

wherein after the application of a voltage to the element, the element is maintained under a reduced pressure of at most 160 Torr;

wherein the organic electrolyte contains

a) a solvent selected from the group consisting of ethylene carbonate, propylene carbonate, butylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, acetonitrile, glutaronitrile and a mixture thereof; or

b) a solvent mixture of sulfolane and a solvent selected from the group consisting of ethylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, acetonitrile, glutaronitrile and a mixture thereof; or

c) a solvent mixture of a sulfolan derivative and a solvent selected from the group consisting of ethylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate, acetonitrile, glutaronitrile and a mixture thereof.

Thus, a high capacity of the electric capacitor is maintained by the claimed methods. As a result of the above methods, a water content within micropores of the capacitor is expelled out into the electrolyte by using benzene or its chlorine derivatives. A preliminary voltage is applied to the electrolyte to gasify the water content by electrolysis. The generated gas is expelled out of the capacitor by applying a vacuum (maintaining the element at a reduced pressure). As a result, a high degree of dehydration of the capacitor is achieved.

Morimoto et al fail to disclose or suggest a method for producing an electric double layer capacitor having 1) the claimed organic solvent in the organic electrolyte, 2) the claimed specific surface area of 100 to 3000 m²/g of the carbonaceous material of the electrodes, and 3) which is maintained at reduced pressure after the voltage is applied as set forth in Claims 5, 12 and 20.

Morimoto et al disclose at column 2, lines 49 to 66 that the low temperature properties of an electric double layer capacitor are improved by adding chlorobenzene having a low solidification point. On the other hand, in the present invention water is removed out of

the capacitor by utilizing the high affinity between benzene or its chlorine derivatives and a pseudo-graphite surface present on pore internal walls of the carbonaceous material, thereby substituting the water content within the micropores with benzene or its chlorine derivatives and expelling the water as described above. Thus, usage of chlorobenzene in Morimoto et al is essentially different from that in the present invention.

Also, the application of voltage disclosed at column 4, lines 40 to 45 was carried out as a reliability test to confirm durability of a capacitor **after** it is prepared. However, in the present invention, the application of voltage is part of the method of producing the capacitor. Furthermore, the present invention includes gasifying of the water content in the electrolyte by electrolysis and subjecting the capacitor to reduced pressure treatment to remove the resulting gas out of the capacitor. As a result, the internal cell pressure is not raised in the capacitor obtained by the present invention, and deterioration of the capacitor performance due to gas remaining within micropores of the electrode can be avoided. In Morimoto et al however, either the water stays in the micropores or if replaced by the chlorobenzene, the water remains in the electrolyte. Upon application of voltage, this water will hydrolyze. However, the generated gas is not removed because the pressure is not reduced. Thus, water in the electrode or the electrolyte or gas in the electrode or electrolyte of Morimoto et al will lead to quick deterioration of the performance of the capacitor. However, this is exactly what the method of the present invention prevents.

Moreover, a comparison of Examples 1 and 3 in Tables 1 and 2 of the specification shows the superior results of the method of the present invention. In Example 1, the pressure is **not** reduced after applying a voltage. As a result, the capacity is reduced in Example 1 after a reliability test, and the resistance increases 20% more than in Example 3 in which the pressure is reduced after application of the voltage (present invention). Thus, the capacitor

produced according to the present invention is superior to a capacitor according to Morimoto et al. This is an unexpected effect. Thus, the present invention is not obvious over Morimoto et al.

In addition, this reference fails to disclose or suggest the claimed solvent combinations. Morimoto et al disclose an electric double layer capacitor having an electrolyte dissolved in sulfolane or a sulfolane derivative (abstract). The sulfolane solvent may be mixed with propylene carbonate or butylene carbonate (Morimoto et al, col. 2, lines 34 and 35). However, the claimed combinations a)-c) of the solvents are neither disclosed nor suggested.

Wei et al do not cure the defects of the primary reference because they fail to disclose or suggest the benzene or its chlorine derivative in the electrolyte or selectively expelling a water content within micropores into the electrolyte as claimed. Furthermore, since Morimoto et al do not remove water within a capacitor cell, there is no motivation to incorporate steps of application of voltage and reduced pressure treatment disclosed in Wei et al into Morimoto et al.

Moreover, Wei et al disclose at column 8, lines 12 to 15 that when a high initial voltage is applied, gas is rapidly generated and the **volume is so increased as to bring a danger of destroying the cell**. Thus, this reference teaches away from continuous application of voltage. Rather, there is a teaching that the voltage should be applied step by step or in a cyclic manner. However, when applying the voltage cyclically, even if the water content in an electrolyte is electrolyzed, there is a possibility that the gas is **redissolved** in the electrolyte **by the increase of the internal pressure**. Thus, the degree of electrolysis of the water content in Wei et al is much lower than in the Examples of the present invention in which a high voltage of 2.8V is **continuously** applied for 12 hours. Thus, even if the

application of voltage and reduced pressure removal disclosed in Wei et al are combined with Morimoto et al, it is impossible to the same degree of dehydration as in the present invention. Thus, the present invention is not obvious over Morimoto et al and Wei et al.

Furthermore, as stated above, if no benzene or chlorine derivative are used, at least some gas generated by the electrolysis of water in the fine pores will also remain in the fine pores thereby deteriorating the performance of the capacitor, particularly after long-term use (specification, page 6, line 7 to page 7, line 12). For example, Example 1 of the present invention demonstrates how a capacitor having benzene performs while Example 13 shows a capacitor having no additive (Table 1 at page 17 and Table 2 at page 18). After a durability test, the service capacitance of Example 1, having the benzene, is 1430 F and has changed from the initial state by 17%, the internal resistance has increased 80% to 5mΩ. The conventional capacitor having no benzene (comparable to Wei et al) has service capacitance of 950 F which correlates to a change from the initial state by 45%, the internal resistance has increased 192% to 8.8mΩ. Thus, an electrolyte having an additive is superior to an electrolyte having no additive, as is further shown by Examples 2-12 at page 17 and 18 of the specification. Further, one of ordinary skill in the art is not likely to consult the Wei et al reference because it pertains to the conventional electric double layer capacitor that has no benzene or chlorine derivative.

Further, Wei et al fails to disclose or suggest the solvent combination in parts b) and c) of Claims 5, 12 and 20.

Therefore, the rejection of Claims 2, 4, 5, 7, 9-11, 20-21, 23, 24 and 27 under 35 U.S.C. §103(a) over Morimoto et al (U.S. 4,725,927) in view of Wei et al (U.S. 6, 152, 970) is believed to be unsustainable as the present invention is neither anticipated nor obvious and withdrawal of this rejection is respectfully requested.

The rejection of Claims 3, 12-15, 17-19 and 22 under 35 U.S.C. §103(a) over Morimoto et al (U.S. 4,725,927) in view of Wei et al (U.S. 6, 152, 970) in further view of Tsushima (JP 100041199) is respectfully traversed.

Tsushima also fails to cure the defects of the primary reference because it fails to disclose or suggest to maintain the element under reduced pressure after the application of the voltage to the element. However, such procedure is advantageous because the generated gas is more effectively removed from the cell (specification, page 8, lines 12-18). In addition, this reference fails to disclose or suggest that the electrolyte has a benzene or its chlorine derivative or a solvent as claimed in Claims 5, 12 and 20 of the present invention.

In addition, there is no disclosure or suggestion that the voltage is applied to the element in a dry atmosphere in an open condition as set forth in Claim 12. Tsushima merely discloses that the voltage is applied before and after the case is sealed (abstract). However, there is no teaching in the reference that the environmental atmosphere be dry.

Therefore, the rejection of Claims 3, 12-15, 17-19 and 22 under 35 U.S.C. §103(a) over Morimoto et al (U.S. 4,725,927) in view of Wei et al (U.S. 6, 152, 970) in further view of Tsushima (JP 100041199) is believed to be unsustainable as the present invention is neither anticipated nor obvious and withdrawal of this rejection is respectfully requested.

The rejection of Claims 8 and 25 under 35 U.S.C. §103(a) over Morimoto et al (U.S. 4,725,927) in view of Wei et al (U.S. 6, 152, 970) in further view of Grigortchak et al (U.S. 5,351,164) is respectfully traversed.

Claims 8 and 25 depend on Claims 5 and 20. Grigortchak et al fail to disclose the claimed methods for producing an electric double layer capacitor having the claimed organic solvent in the organic electrolyte.

Grigortchak et al does not at all pertain to an electric double layer capacitor having an

organic electrolyte as claimed. This reference discloses that the electrolyte consists of an **aqueous solution** of alkali, preferably LiClO₄, in propylene carbonate (Grigortchak et al, col. 6, lines 59-61). Thus, one of skill in the art has no motivation whatsoever to consult this reference as it pertains to a completely different type of capacitor. Looking at a capacitor that actually utilizes an aqueous electrolyte is **contrary to the present invention**, where the presence of water is to be excluded from the organic electrolyte.

Therefore, the rejection of Claims 8 and 25 under 35 U.S.C. §103(a) over Morimoto et al (U.S. 4,725,927) in view of Wei et al (U.S. 6, 152, 970) in further view of Grigortchak et al (U.S. 5,351,164) is believed to be unsustainable as the present invention is neither anticipated nor obvious and withdrawal of this rejection is respectfully requested.

The rejection of Claim 16 under 35 U.S.C. §103(a) over Morimoto et al (U.S. 4,725,927) in view of Wei et al (U.S. 6, 152, 970) and Tsushima (JP 100041199) in further view of Grigortchak et al (U.S. 5,351,164) is respectfully traversed.

Claim 16 depends on Claim 12. None of the cited references discloses or suggests a method for producing an electric double layer capacitor having the claimed organic solvent in the organic electrolyte as discussed above. Even a combination of Morimoto et al, Wei et al, Tsushima and Grigortchak et al does not result in the invention as claimed in Claim 16.

Therefore, the rejection of Claim 16 under 35 U.S.C. §103(a) over Morimoto et al (U.S. 4,725,927) in view of Wei et al (U.S. 6, 152, 970) and Tsushima (JP 100041199) in further view of Grigortchak et al (U.S. 5,351,164) is believed to be unsustainable as the present invention is neither anticipated nor obvious and withdrawal of this rejection is respectfully requested.

In regard to the Examiner's request to point out the inventor and invention dates of each claim that was not commonly owned at the time a later invention was made, Applicants'

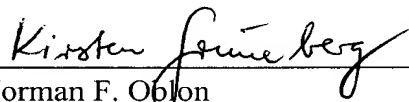
Representative is confirming with the Applicants that all claims were commonly owned.

Applicants' Representative will update the Examiner in the event that the claims were not commonly owned.

This application is now in condition for allowance. Early notice of such action is earnestly solicited.

Respectfully submitted,

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AMENDMENT AND REQUEST FOR RECONSIDERATION

IN THE SPECIFICATION

Page 18, please amend Table 2 as follows:

	Initial stage		After the durability test			
	Service capaci- tance	Internal resis- ance	Service capaci- tance	Change in capaci- tance	Internal resist- ance	Change in [capaci- tance] <u>resistance</u>
	(F)	(mΩ)	(F)	(%)	(mΩ)	(%)
Ex. 1	1720	2.8	1430	-17	5.0	+80
Ex. 2	1700	2.7	1450	-15	4.5	+65
Ex. 3	1710	2.7	1470	-14	4.3	+60
Ex. 4	1720	2.9	1440	-15	4.7	+62
Ex. 5	1710	3.0	1400	-16	5.5	+62
Ex. 6	1730	2.5	1440	-17	4.8	+79
Ex. 7	1710	2.6	1440	-16	4.3	+66
Ex. 8	1710	2.6	1470	-14	4.2	+62
Ex. 9	1710	2.8	1500	-12	4.4	+58
Ex. 10	1720	2.8	1480	-14	4.5	+61
Ex. 11	1710	2.7	1490	-19	4.3	+59
Ex. 12	1700	2.7	1280	-19	5.5	+105
Ex. 13	1730	3.0	950	-45	8.8	+192
Ex. 14	1720	2.9	480	-72	18.9	+520
Ex. 15	1710	2.8	1520	-11	4.3	+55
Ex. 16	1720	2.7	1530	-11	4.2	+54